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This work was accomplished primarily by Allison G. Wozniak, a graduate research assistant who has completed the Master of Science in Meteorology program at the South Dakota School of Mines and Technology. Ms. Wozniak was guided and assisted in her work by L. R. Johnson and the principal investigator. Invaluable guidance was supplied by Dr. James Holdeman, NASA Lewis, the manager of the Global Atmospheric Sampling Program (GASP). Dr. Gregory Nastrom, St. Cloud (Minnesota) State University, who has used the GASP data set to provide unique views of the distribution of ozone, clouds, and atmospheric waves and turbulence, in the upper troposphere/lower stratosphere region, was also extremely helpful. Finally, Dr. Terry Deshler, University of Wyoming, supplied observations from the university's upper atmospheric monitoring program for comparison to GASP data.

We analyzed all of the ambient aerosol, ozone, and carbon monoxide data acquired during GASP, which was conducted from 1975-1979. (Proposed analysis of some cloud microphysical characteristics was eliminated as the scope of the funding was reduced from that originally proposed.) Sampling during this program was conducted using automated instrumentation installed on commercial airliners in routine service, providing good coverage along the major air routes through the upper troposphere and lower stratosphere over what was, in that era, known as the "non-Communist world". The analysis consisted of three phases, including review of relevant technical reports and previously-published work based on GASP, detailed analysis of data from selected flights, and exploratory analysis of the entire data set.

Review of technical reports and prior publications suggests that the ozone data from GASP are reasonably good. Ozone data are available from 1975-1979. The relatively high data quality is due to a combination of factors, including mature instrumentation technology, rapid attention to some problems that developed in the sampling system leading to ozone destruction in the system, and careful attention to periodic refurbishment and calibration of instruments in service. An exhaustive survey of GASP carbon monoxide data (available from 1977 onward), by M.-F. Wu and R. E. Newell (Wu, 1981) shows that these data were fairly noisy. This is due to an immature instrumentation technology, and problems in the scrubbers used to exclude water from the air sample entering the instrument. Laboratory work with the optical particle counters used in GASP (with data reported beginning in 1977) suggests that there are relatively large uncertainties in the instrument sample volume, and some uncertainties in particle sizing, due to non-uniform illumination of the sample volume. These counters were state-of-the-art for their time, but relatively crude by today's standards. During GASP and in subsequent analyses, these counters were used to indicate when an aircraft was in-cloud. Little else has been done with their data. Finally, very little attention has been devoted to condensation nucleus measurements (available beginning in late 1977) in these reports and publications, but the information available suggests that the data should be reasonably good.

Based on this review, and with the objectives of the AEAP in mind, we concentrated our analyses in three areas. The first was to extend prior analyses of ozone observations. The second was to evaluate the data from the optical particle counters in order to determine to what population of aerosol particles they were responding, and to determine if there were discernible effects of aircraft emissions on the geographical and meteorological distributions of these particles. Finally, we evaluated and analyzed the condensation nuclei data in order to better understand their quality and, again, to look for possible impact of aircraft emissions on their distribution.

In order to evaluate data quality, we analyzed in detail the data from several hundred individual flights. Archived upper-air meteorological data were included in the analysis, in order to better understand typical variations of the constituents with meteorological situation. As expected, ozone mixing ratios increased dramatically between the upper troposphere and lower stratosphere. Condensation nuclei and carbon monoxide mixing ratios were generally lower in the stratosphere than in the troposphere. The optical particle counter, from which particle concentrations were sized in 3 cumulative size categories, indicated higher particle concentrations for all size categories in clouds, few giant particles (particles larger than 3  $\mu\text{m}$  diameter) outside of clouds, and an increase in concentration in the large particle category (particles larger than 1.4  $\mu\text{m}$  diameter) with distance above the tropopause.

Contemporaneous measurements of condensation nuclei and light-scattering particles were obtained from several routine vertical soundings conducted with balloon-borne instrumentation packages as part of the ongoing University of Wyoming monitoring program. Several days were found when GASP-instrumented aircraft traversed the same region sampled by one of these balloon packages within a few hours of the balloon sounding. Data from these days showed that the GASP condensation nuclei instruments indicated much lower concentrations ( $\sim 1/5$  X) compared to the Wyoming instruments. The GASP light-scattering particle counter, on the other hand, indicated much higher concentrations (10's X to 100's X) than the corresponding Wyoming counter.

Further study of the published literature on condensation nuclei measurements, and discussion with others who make these observations, leads us to conclude that the difference in the GASP and Wyoming concentrations is due to the differences in measurement technology between the two instrumentation packages. The expansion/condensation counters with water as a condensing vapor, used in the GASP packages, yielded concentrations within the range expected based on contemporaneous measurements made using similar instruments in the upper troposphere/lower stratosphere. On the other hand, the Wyoming counter was based on a diffusion chamber in which glycol was condensed on particles in order to render them detectable. This was a new technology in that era. The range of both the Wyoming measurements of that era and those made currently using similar technology are also consistent. We propose that the difference in response of counters based on the two different technologies is due to two factors. First, a significant fraction of the condensation nucleus population may be volatilized in the sampling train of the GASP system and any system in which air is compressed before entering the instrument. Second, more particles may act as condensation nuclei to the organic vapors used in the diffusion counters, than to water which is condensed in the expansion counters.

The disparity in indicated concentrations of light-scattering particles is harder to explain. It could be due to a combination of both sizing and concentration errors. The disagreement in concentration is far outside the range of uncertainty indicated in the published reports from GASP (e.g. Nastrom et al, 1981), which is based on laboratory work conducted during GASP. Further investigation as part of our work has shown that, in the few cases where different GASP instruments sampled over the same region within the same 3-month meteorological season, the range of concentrations reported by the different instruments is similar, indicating that within the GASP data set, different instruments had similar responses. Given the dramatic disagreement between the GASP and Wyoming instruments, we conducted additional analyses using the GASP data to try to identify the particle size range to which these counters were responding. The results are discussed below.

In our analyses, we focused on the distribution of the gases and particles relative to the tropopause (taken from contemporaneous operational analyses by the U. S. National Meteorological Center), relative to geographic regions, and relative to meteorological seasons. The results are presented in Wozniak (1997). Ozone mixing ratio, in both the troposphere and stratosphere, was observed to vary with time and season in accord with other published analyses of observations from that era. Springtime peaks were observed in most regions, and there was an overall upward trend in concentration during the era 1975-1979. Ozone increased above the tropopause, with a typical mixing ratio near the tropopause of 100 ppbv and increases with altitude of the order of 100 ppbv/km above the tropopause. The "natural" variation with season and altitude is so large as to make it impossible to discern any evidence of regional variations that might be due to regional variation in aircraft emissions, which are expected to be of the order of 1% (Friedl et al, 1997).

The carbon monoxide data were so noisy as to preclude the possibility of analyzing for trends with time or season within the level of effort funded. Typically, the "believable" carbon monoxide mixing ratios decreased with distance above the tropopause. Typical upper tropospheric mixing ratios were in the range 50-100 ppbv varying little with altitude, while most stratospheric reports were less than 60 ppbv. Further analysis would be feasible, although somewhat laborious, with guidance from Wu (1981) as to which data are good and which are not.

The total concentration of light-scattering particles, diameter  $> 0.9 \mu\text{m}$ , appears to be strongly influenced by instrument noise. This parameter varies little with altitude, region, season, when an aircraft crosses a meteorological boundary such as the tropopause, a jet stream axis, etc., or when the aircraft encounters a cloud. The number mixing ratio of particles with diameter  $> 1.4 \mu\text{m}$  in clear air generally correlates well with ozone mixing ratio and increases with distance above the tropopause. We interpret this to indicate that this size category represents the aged stratospheric sulfate aerosol, probably the upper end of its size distribution. Only relative variations could be analyzed. No absolute calibration of concentrations is possible. The regional variation of this particle population is similar to that of ozone, and does not vary consistently between heavily- and lightly-traveled air traffic regions. Finally, as reported in other GASP literature (e.g. Nastrom et al, 1981), reports of significant concentrations of particles with diameter  $> 3 \mu\text{m}$  indicate that the aircraft is in cloud. When the aircraft is in-cloud, the concentration of particles with diameter  $> 1.4 \mu\text{m}$  nearly equals the concentration of particles with diameter  $> 3.0 \mu\text{m}$ .

The condensation nuclei number mixing ratio, alone among the observations studied, may indicate an effect due to aircraft emission. These mixing ratios are generally higher at altitudes and in regions with heavier air traffic. However, they are also higher in cloudy than in clear air, and the more-heavily-traveled air routes across the North Atlantic and continental US are in general cloudier than some other regions analyzed. We are currently in the process of separating observations between clear and cloud regions and re-analyzing the variation with altitude and region using only clear-air observations.

Although the work under this grant has ended, we will publish the main results of our work in the near future using local funds, after completing analyses of condensation nuclei concentrations in which only clear-air observations are used.

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# Exploratory Analysis of Global Atmospheric Sampling Program Data

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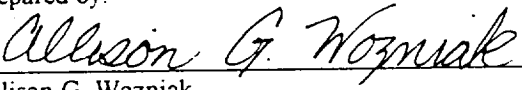
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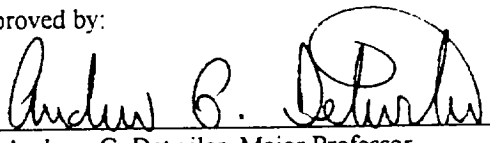
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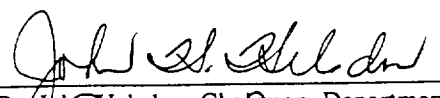
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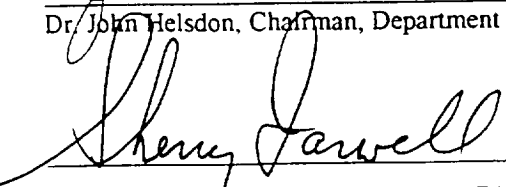
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## ABSTRACT

The NASA Global Atmospheric Sampling Program (GASP) was an initial attempt to use commercial airliners to collect automated atmospheric measurements. Measurements were taken from March 1975 through June 1979. The parameters that were gathered include: aircraft location information, meteorological variables, and atmospheric trace constituents including ozone, light-scattering particles, condensation nuclei, and carbon monoxide. As part of the subsonic assessment in NASA's Atmospheric Effects of Aviation Pollution project, the GASP measurements are being evaluated to determine if they are of sufficient quality for use as baseline measurements from the late 1970's.

In an effort to investigate data quality, numerous flight records were examined. Some flight records revealed excessively high carbon monoxide records on the order of several hundreds ppbv. Significant concentrations of particles with  $D > 3.0 \mu\text{m}$  only occurred in-cloud. Additionally, particle data from the University of Wyoming (UW) balloon soundings were compared to the particle and condensation nuclei data from GASP. When compared to the UW data, the GASP CN readings were an order of magnitude lower and optical particle counts were  $\sim 1.5$  orders of magnitude higher. Due to discrepancies between GASP and UW data, the GASP data are not suitable for direct comparisons to current measurements. However, the data are suitable for evaluating relative regional differences of these parameters. With further research, the data could be used indirectly to establish baselines by scaling GASP data to a long running data source, like the UW balloon data, through the entire data set.

The analysis of ozone concentration versus offset from tropopause shows a distinct increase in slope above the tropopause. The seasonal average of ozone concentration shows an annual fluctuation peaking in spring for most regions. An increase in ozone occurs during the four year data period due to natural forcings. The average vertical profiles for the northern and southern hemispheres show similar ozone averages around 50 ppbv in the upper troposphere to several hundreds ppbv in the lower stratosphere.

The particles with  $D > 1.4 \mu\text{m}$  and condensation nuclei concentrations show a seasonal cycle with a spring/summer peak. The  $D > 1.4 \mu\text{m}$  particles show an increase in concentration with height, which would correspond to a stratospheric source for these particles. Just below the tropopause, average concentrations of particles with  $D > 1.4 \mu\text{m}$  are higher in the northern hemisphere ( $\sim 82700/\text{m}^3$ ) than the southern hemisphere ( $\sim 12800/\text{m}^3$ ), while the rest of the particle concentration vertical profile for the hemispheres is similar. Condensation nuclei concentrations are negatively correlated with height, which is consistent with a ground source for these particles. In the northern hemisphere, tropospheric condensation nuclei concentrations average  $\sim 200/\text{cm}^3$  while the stratospheric concentration is  $\sim 60/\text{cm}^3$ . Carbon monoxide concentrations show the same inverse relationship with height as the condensation nuclei, despite significant instrument noise.